Determination of Exchange Parameters from Magnetic Susceptibility

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Abstract

We report a novel practical method to determine exchange parameters by using experimental susceptibility data in a relatively narrow temperature region and a low order high-temperature-expansion equation. This method is applied to a square lattice and a CaV_4O_9-type lattice, and its accuracy is discussed.

Keywords: magnetic susceptibility, exchange parameter, high temperature expansion, Heisenberg model, square lattice, CaV_4O_9

When we describe magnetic properties of a matter by a Heisenberg model, we must determine exchange parameters in the model to fit the matter. It is sometimes performed by the high temperature expansion (HTE) of the susceptibility of the Heisenberg model and by the comparison of it to the experimental data. However meaningful experimental data are often restricted in a low temperature region because of, *e.g.*, the temperature dependence of the lattice constant or structure. Further it becomes hard to calculate high order terms of the HTE because of striking increase of the number of diagrams if the model includes many exchange parameters. In some of such cases, an ordinary method of the direct fitting may not estimate exchange parameters with sufficient accuracy.

In this article, we report a novel practical method to determine exchange parameters by using experimental susceptibility data in a relatively narrow temperature region and a low order HTE equation. The essence of this method is that we construct an experimental formula for the experimental susceptibility

data in a power series of 1/T and then compare the coefficients of this formula to those of the HTE equation derived from the assumed Heisenberg model.

We denote the experimental data for the magnetic susceptibility by $\chi^{EXP}(T)$ and the experimental formula in a power series of 1/T by $\chi(T)$. To make expansion coefficients dimensionless we use $x = T_0/T$ as the expansion parameter, where T_0 is a constant temperature chosen arbitrarily. Then $\chi(T)$ is written as

$$\begin{aligned} \chi(T) &= \lim_{n\to\infty} \chi^{(n)}(T), \\ \chi^{(n)}(T) &= \frac{C}{T}\sum_{m=0}^{n} A_m x^m, \end{aligned} \tag{1}$$

where C is Curie constant determined by experiments. The zeroth expansion coefficient is set as $A_0 = 1$ and the others (A_m's for $m \geq 1$) are fitting parameters to be determined.

Using the same coefficients A_m's, we define the following quantity:

$$\phi_m(x) = A_m + \sum_{l=1}^{\infty} A_{m+l} x^l \tag{2}$$

for each m. In the high temperature limit, it reduces to a coefficient as

$$\phi_m(0) = A_m. \tag{3}$$

This quantity $\phi_m(x)$ is also defined by the recursion equation:

$$\begin{aligned} \phi_0(x) &= \frac{T}{C}\,\chi(T), \\ \phi_m(x) &= (\phi_{m-1}(x) - A_{m-1})\frac{1}{x} \end{aligned} \tag{4}$$

for positive integer m.

The experimental data corresponding to $\phi_m(x)$ is similarly defined in the same recursive transformation:

$$\begin{aligned} \phi_0^{EXP}(x) &= \frac{T}{C}\,\chi^{EXP}(T), \\ \phi_m^{EXP}(x) &= (\phi_{m-1}^{EXP}(x) - A_{m-1})\frac{1}{x}. \end{aligned} \tag{5}$$

The quantity ϕ_m^{EXP} is actually determined only when A_{m-1} is known. It is also noticed that ϕ_m^{EXP} is defined only at experimental data points in $\{x\}$; e. g. it is not defined at very high temperatures ($x \sim 0$). ϕ_m must fit ϕ_m^{EXP}, since χ is constructed to fit χ^{EXP} and ϕ_m is produced by the same transformation as ϕ_m^{EXP}.

In the following, we inductively determine $\{A_m\}$ together with $\{\phi_m^{EXP}(x)\}$. For $m = 0$, $\phi_0^{EXP}(x)$ is defined by eq. (5) and $A_0 = 1$ is a definition. Assuming

that $\phi_{m-1}(x)$ and A_{m-1} are known, we construct $\phi_m^{EXP}(x)$ and A_m. $\phi_m^{EXP}(x)$ is simply given by eq. (5). To determine A_m, we consider the high temperature limit ($x \to 0$) for $\phi_m^{EXP}(x)$ which corresponds to eq. (3) for $\phi_m(x)$. Since $\phi_m^{EXP}(x)$ does not have data points in the vicinity of x=0, we extrapolate the graph of $\phi_m^{EXP}(x)$ to $x = 0$ by using a polynomial fitting function:

$$f_m(x) = \sum_{l=0}^{L} a_{ml} x^l, \tag{6}$$

where coefficients a_{ml}'s ($l = 0, \cdots, L$) are fitting parameters to be determined for each m. The order L of the polynomial is empirically chosen as 8 to 10 and is inspected with some examples as mentioned later. A_m is then determined by $A_m \simeq f_m(0) = a_{m0}$ corresponding to eq. (3). Thus we have completed the inductive definition and have constructed an experimental HTE formula (1b).

On the other hand, we start from a Heisenberg Hamiltonian H which is assumed to describe the magnetic properties of the material. The assumed Hamiltonian includes a set of exchange parameters $\{J_i\}$. For this Hamiltonian, we calculate the theoretical HTE formula

$$\chi^{HTE}(T) = \frac{C}{T} \sum_{m=0}^{\infty} F_m x^m \tag{7}$$

by the standard diagrammatic method. [1] The expansion coefficients F_m's are functions of the set of exchange parameters $\{J_i\}$.

Lastly we compare the theoretical HTE formula (7) to the experimental formula (1). If the assumed Hamiltonian exactly describes the material, both the coefficients must be the same. Hence we can determine the exchange parameters by solving the set of equations $\{F_m(\{J_i\}) = A_m\}$. In real cases, however, the model Hamiltonian may not perfectly describe the matter and the experimental data may include some errors. We usually must be satisfied with a set $\{J_i\}$ which gives approximate matching between $\{F_m\}$ and $\{A_m\}$.

We inspect the method mentioned above for the Heisenberg model in a simple square lattice. Only an exchange parameter J is between nearest neighbor spins. As data for susceptibility, we use the result of Quantum Monte Carlo (QMC) simulation by Troyer et al. [2] instead of data of a real experiment. We choose the expansion parameter as $x = 1/T$ in the unit of $T_0 = 1$ and use 12 QMC data points in $0.05 \leq T \leq 1.5$. The data is for $J = 1$ and $C = 0.25$.

Following the above procedure, we obtained coefficients of HTE as $A_1 = -0.994$, $A_2 = 0.483$ and $A_3 = -0.135$ for $L = 8$. These values are little changed when the order L of the fitting function changes from 6 to 10. We show $\phi_m^{EXP}(x)$ along with $f_m(x)$ for $m = 1$ to 3 in Fig. 1.

The corresponding theoretical coefficients are calculated as $F_1 = -J$, $F_2 = J^2/2$ and $F_3 = -J^3/6$. In this one-parameter case, equation $F_1(J) = A_1$ is sufficient to determine J. The solution is $J = 0.994$, which is close to the

Figure 1: Transformed QMC data $\phi_m^{EXP}(x)$ (circle) and fitting function $f_m(x)$ (line) with $m = 1$ to 3 for a square lattice.

exact value $J = 1$. We estimate the accuracy of A_m by the deviation $d_m = 2|(A_m - F_m(1))/(A_m + F_m(1))|$. It is given as $d_m = 0.006$, 0.017 and 0.070 for $m = 1$, 2 and 3, respectively. This result, i. e. this method, is fairly accurate despite the small number of the data points.

We also carried out the same analysis by restricting the temperature region as $0.05 \leq T \leq 0.9$, which includes only 9 data points. This region is below the maximum point of $\chi^{EXP}(T)$ and is rather a low temperature region. The present method gives $A_1 = -1.019$, $A_2 = 0.541$ and $A_3 = -0.185$ for $L = 8$. The corresponding deviations are $d_m = 0.019$, 0.079 and 0.327 for $m = 1$, 2 and 3, respectively. This estimation is practically sufficient to determine J from A_1.

Next we apply our method to a Heisenberg model on a lattice which is a simplified two-dimensional model used for CaV_4O_9. This model includes two exchange parameters, J_0 for the plaquette link and J_1 for the dimer link. Theoretical coefficients are obtained as $F_1(J_0, J_1) = -(2J_0 + J_1)/4$ and $F_2(J_0, J_1) = (4J_0 - J_1)J_1/16$. Instead of experimental data, we use QMC data [2] and apply our method to determine J_0 and J_1. We used 15 QMC data points in $0.075 \leq T \leq 1.5$ and the data are for $C = 0.25$ in the unit of $T_0 = 1$. We examine two typical cases of J_0/J_1 =1.0 and $J_0/J_1 = 0.5$. The system has no spin gap in the former case, while it has in the latter. [2]

For the data of $(J_0, J_1) = (1, 1)$, our method produces coefficients, $A_1 = -0.750$ and $A_2 = 0.166$. The fitting was done by the 8th order polynomial ($L = 8$), f_m's. The corresponding exact values are $F_1 = -0.750$ and $F_2 = 0.188$, and the deviations are $d_1 = 0.00$ and $d_2 = 0.12$. The complete matching between A_1 and F_1 over 3 digits are occasional, since the last digit changes if we change L from 8 to 9 or 10. We calculated the exchange parameters as $(J_0, J_1) = (0.83, 1.34)$ or $(1.17, 0.66)$ by solving $F_1(J_0, J_1) = A_1$ and $F_2(J_0, J_1) = A_2$.

Similarly we applied our method for the data of $(J_0, J_1) = (0.5, 1)$. We obtained HTE coefficients as $A_1 = -0.500$ and $A_2 = 0.052$. The corresponding exact values are $F_1 = -0.500$ and $F_2 = 0.063$, and the deviations are $d_1 = 0.00$ and $d_2 = 0.19$. The obtained exchange parameters are $(J_0, J_1) = (0.46, 1.08)$.

The estimation gives better result for a square lattice than for a CaV_4O_9 lattice. A reason may be small HTE coefficients included in the latter lattice. Such small coefficients seems to appear, when the coordination number is small so that the spin fluctuation is strong. Large fluctuation is also a cause of the spin-gap formation. The present method is applicable to gapful systems as well as gapless systems as long as we only need coefficients of low orders.

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